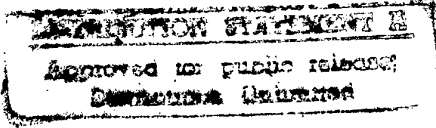


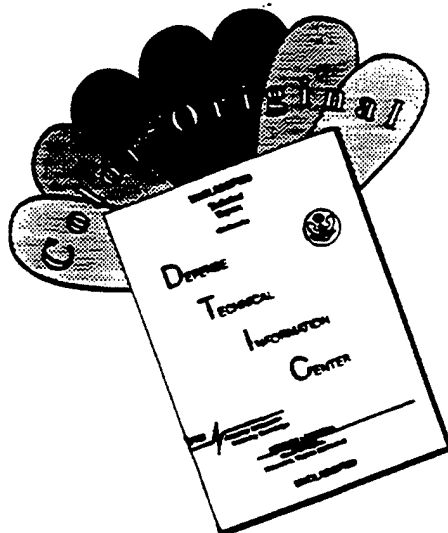
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BACKSCATTER AND TRANSMISSION OF AEROSOL AT UV THROUGH MIDDLE IR WAVELENGTHS

ABSTRACT

Simultaneous direct measurements of volume extinction coefficient, σ_e , and volume backscatter coefficient, σ_b , for obscuring aerosol of carbon graphite flakes have been carried out in the laboratory at four wavelengths: 1064, 532, 355 and 266 nm. Despite the low backscattered signal, reasonably good agreement was obtained between the experimentally determined values for σ_e/σ_b and the predicted values. The results are consistent with the assumption that the carbon aerosol flakes possess shapes between those of spheres and infinite thin parallel slabs or large thin disks. Forward scattering measurements are obtained at wavelengths 1064 and 532 nm for water droplet polydispersions and compare favourably with theoretical prediction for relatively narrow size distributions. However, for broader size distributions, the contribution by forward scattered radiation to the estimated signal is significant and needs to be accounted for in extinction and backscatter measurements.

Background aerosol particle and mass distribution measurements of the ambient natural aerosol were carried out at the Mace Head atmospheric research field station near Carna, Co. Galway (53°19'N, 9°54'W) on the west coast of Ireland. Aerosol particle volatility was also measured which yielded information on aerosol chemical constituents. Aerosol microphysical and physico-chemical data is obtained for background marine air masses and also for polluted air for both summer and winter periods. Background marine aerosol accumulation mode particle number concentration range in value between around 40 to 200 cm^{-3} . The use of a low pressure multi-stage impactor permitted the measurement of the background aerosol mass distribution which yielded accumulation mode aerosol mass of about 5.5 $\mu\text{g m}^{-3}$.

A passive biological aerosol sampler, the Tauber trap, and a continuous (with a four hour resolution) Buckard volumetric spore trap were used for the collection and measurement of pollen and spore species. Sample preparation and acetolysis of the samples is described in the 7th interim report. Relative pollen concentration in terms of number of pollen grains per unit time per cross-sectional area of the Tauber trap inlet

were obtained for a range of sampling sites (including Mace Head) in the west of Ireland over the period November 1992 through June 1994. Absolute pollen and spore number concentration were also measured at the Mace Head field site for the period March 1993 through June 1994, and ranged in value from zero up to around 700 counts m^{-3} , with most measurements $< 200 \text{ m}^{-3}$. Pollen grain and spore concentration m^{-3} were obtained also for individual taxa (pollen and spore species). Comparison of the two sampling methods is also discussed. Some preliminary work and review of glass cyclone bioaerosol samplers to obtain ambient samples for the determination of the total fluorescence of the biological background aerosol is also presented.

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Backscatter and Transmission of Aerosol at UV Through Middle IR Wavelengths

1. INTRODUCTION

The work carried out under this proposal encompassed a range of scientific investigations. These included the following areas of work:

- (a) Backscatter and transmission of black carbon aerosol in the laboratory.
- (b) Field measurements of the natural background aerosol made at the remote field station at Mace Head on the west coast of Ireland.
- (c) Volatile properties of black carbon, measured in the laboratory and in the field.
- (d) Measurements of the biological aerosol at a range of sites in the west of Ireland, with particular emphasis on pollen aerosol.

2. EXPERIMENTAL APPARATUS AND TECHNIQUES

2.1 Backscatter and Transmission Instrumentation and Experimental Procedures

A Continuum Surelite 10 Hz Nd:Yag laser which included 2nd, 3rd and 4th harmonics was used to carry out transmission and backscatter measurements in the laboratory. Beam profiles were also obtained.

Pyroelectric probes from Molelectron were used to detect the transmitted and backscattered energies from the Continuum Surelite Nd:Yag pulsed laser system. The nominal maximum energies of the Continuum Surelite laser are 365 nJ at the fundamental wavelength of 1064 nm, 165 mJ at the second harmonic wavelength of 532 nm, 55 mJ at the third and fourth harmonic wavelengths of 355 and 266 nm. The pyroelectric probes are capable of withstanding the energy densities and have fast response times (up to 50 pulses per second) enabling single or multiple laser shots at 10 Hz to be used. This is a considerable advantage over the slow response volumetric detection probes which only allowed single shots to be measured.

The J50 probe with beam expander (JBX) enables the higher transmitted energies to be detected in the range μJ to J. The J4-09 probe enables the lower backscattered energies to be measured in the range 50 nJ to 5 mJ.

A dual channel power/energy detector and meter system (JD 2000 from Molelectron) has been calibrated for the two probes to measure the energy (or power) directly (A and B) and also gives direct readings A/B, B/A and the average over 1 to 100 slots (with standard deviation). The ability to use both probes simultaneously (at 10 Hz in present measurements) means that any fluctuations in the aerosol cloud and resulting signals are less significant. The meter is triggered directly from the laser power supply.

A 50 mm diameter circular plano-plano optic (10 mm thickness) made from fused silica with an 8 mm diameter hole bored through it at an angle of 45 degrees centred onto a highly reflecting face is used. Ultra high energy coatings on both surfaces have damage thresholds of 2J in 1 ns at 1064 and 532 nm. The reflecting face has greater than 99.5% reflectance. Hence this mirror is suitable for use with the Continuum Surelite Nd:Yag laser at 1064 and 532 nm.

The laser beam was set up to pass through the mirror hole into the aerosol chamber. The transmitted beam is measured directly by the pyroelectric probe. The back scattered signal is reflected immediately below the mirror hole onto the pyroelectric probe. This set-up has the advantage that this probe is not damaged by the main beam (as it is well away from it) yet can measure backscatter signals at an angle of 0.3° from the far end of the aerosol chamber to 2° from the near end of the aerosol chamber under the present experimental arrangement.

Two black boxes are designed to give multiple (> 20) internal reflections from black matt surfaces to reduce any stray signals from them. One is positioned behind the mirror (opposite side to the probe) and is particularly useful when set up to absorb any clipped signal. The other movable black box at the far end beyond the aerosol chamber is used to check for any stray noise.

2.2 Biological aerosol samplers - Description and Procedures

2.2.1 Tauber Trap

The Tauber trap (Tauber, 1965) is a passive Biological aerosol sampler used for the collection of spore and pollen species. The Tauber traps are changed regularly once a month within a day of each other so as to permit intercomparison of the biological aerosol data for the different sites.

The trap has an aerodynamically shaped top plate, through which the biological aerosol enters. The bottom of the trap is filled with glycerol in order to prevent drying out of the pollen or spores in the event of evaporation. A few grains of thiamine crystals are added to the glycerol to prevent spores developing into fungi. Formaldehyde is also added to deter insects from entering the trap.

After collection of the trap, the first stage of preparation involves each sample having a known quantity of Lycopodium spores added to it in order to determine the actual pollen/spore count of the total sample. The samples obtained for analysis are centrifuged in order to concentrate the sample and then stained by boiling with concentrated sulphuric acid and acetic anhydride. Slides are prepared in the standard way from concentrated solutions. The slides are analysed, which involves counting all of the pollen, spores and lycopodium and recording the values. The counting sheet used for the pollen and spores in the Tauber trap is documented in the 3rd interim report.

2.2.2 Buckard Sampler

A seven day Buckard volumetric spore trap (Jaeger and Mandrioli, 1991) was also used for the measurement of spores and pollen grains at the Mace Head site. Number concentration (m^{-3}) of pollen and spore species are obtained from the mean flow rate of the Buckard sampler. The biological species are collected on melinex tape mounted on a rotatable drum. The wheel in the Buckard volumetric sampler is changed each week at the same time. Slides are made from the melinex tape, which are stained with fushin dye. The biological species are counted and recorded at intervals of two hours on the melinex tape using a light microscope.

2.3 Aerosol Size Distribution and Volatility Instrumentation

The main instruments used for aerosol particle size measurements were the Particle Measuring Systems (PMS) and Active Scattering Aerosol Spectrometer Probes (ASASP-X) using both 64 and 32 channels. The aerosol particles were generally some degrees above ambient temperature before entry into the particle spectrometer since they were passed through a quartz heater tube. Thus it was the dry aerosol fraction which was sampled at the site. The aerosol was drawn through the probe at a flow rate of $1 \text{ cm}^3 \text{ s}^{-1}$. All particles passing through the instrument were counted. The absolute concentrations measured by the ASASP-X were therefore as accurate as the measurement of flow rate through the instrument.

The volatility instrumentation consists of a quartz tube assembly preceding the particle sizing ASASP-X system. This quartz tube is heated to a maximum temperature of 870°C in approximately 3 minutes and then allowed to cool back to near ambient temperature for the remaining 57 minutes of the hour long heating cycle. Further details of the volatility system are given by Jennings and O'Dowd (1990). Ammonium sulphate shows a fairly sharp drop-off in number concentration, particularly for the smaller sized particles, at about 180°C . The marine aerosol number concentration shows a definite fall off, particularly for the largest size range, close to 600°C , which is indicative of the presence of sodium chloride particles. A sharp fall off in particle concentration at around $720\text{--}730^\circ\text{C}$ is due to the presence of elemental carbon (Jennings et al., 1994).

Condensation nuclei (CN) number concentration was measured using a TSI model 3760 condensation particle counter which was modified for use at marine sites (Maring and Schwartz, 1994). Cloud condensation number concentration was determined using a CCN counter manufactured by DH Associates (model M1). This instrument is a static thermal diffusion chamber illuminated by a laser and viewed by a CCD camera. More detail of the CCN system is described by Gras et al. (1996). The winter CCN measurements were taken at 0.3% supersaturation while the summer measurements were at 0.5% supersaturation. A commercial instrument known as an aethalometer (Hansen et al., 1984) was used to obtain the mass concentration of black carbon (BC)

from optical attenuation measurements assuming an attenuation cross section, σ_{Aeth} , value of $19 \text{ m}^2 \text{ g}^{-1}$.

3. RESULTS OF BACKSCATTER AND TRANSMISSION MEASUREMENTS IN LABORATORY GENERATED AEROSOL

3.1 Measurements Using Water Clouds

Simultaneous measurements of volume backscatter coefficient, $\sigma_b \text{ (m}^{-1} \text{ sr}^{-1}\text{)}$ and volume extinction coefficient, $\sigma_e \text{ (m}^{-1}\text{)}$ were obtained in water droplet clouds at wavelength $1.06 \text{ }\mu\text{m}$. A range of water cloud generation methods were used with humidifier and nebuliser systems. It was found that the average $\sigma_e/\sigma_b \text{ (sr)}$ ratio value was 14.1 ± 0.5 which is in fair agreement with that predicted (Pinnick et al., 1983).

Good agreement was obtained between measured and predicted journal scattered signal by a cloud of water droplets at wavelengths 0.532 and $1.064 \text{ }\mu\text{m}$.

3.2 Measurements Using Graphite Powder (Astbury M260)

A special aerosol chamber was designed (detailed in the 4th interim report) for the measurement of transmission and backscatter for obscuring graphite aerosol. Simultaneous direct measurements of volume extinction and backscatter coefficient were carried out for laboratory graphite aerosol at four wavelengths: 1.064 , 0.532 , 0.355 and $0.266 \text{ }\mu\text{m}$. Despite the low backscatter signal, reasonably good agreement was obtained between the experimental values for the σ_e/σ_b and theoretical values (assuming spherical shape), as shown in Table 1. Further discussion of the experimental results is given in the 5th interim report.

Table 1 Extinction and Backscatter from Carbon Graphite Flakes (Asbury M260 #4676)

Wavelength	Refractive Index (a) Real n	(b) Imaginary k	R	σ_e/σ_b sr (theoretical) (a) radius range 0.75 to 10 μm	(b) Geometric mean radius $r_g = 1.2 \mu\text{m}$	σ_e/σ_b sr (experimental)	Number of measurements
266	1.39	0.99	1.6	160 \rightarrow 171	166	126(\pm 17)	74
355	1.57	0.76	2.4	216 \rightarrow 235	225	173(\pm 15)	137
532	1.64	0.84	2.6	185 \rightarrow 215	204	228(\pm 24)	81
1064	1.89	1.27	4.3	109 \rightarrow 141	133	243(\pm 13)	80

4. NATURAL BACKGROUND AEROSOL FIELD MEASUREMENTS AT THE BACKGROUND FIELD STATION AT MACE HEAD, ON THE WEST COAST OF IRELAND

4.1 Aerosol Particle Size Measurements at Mace Head

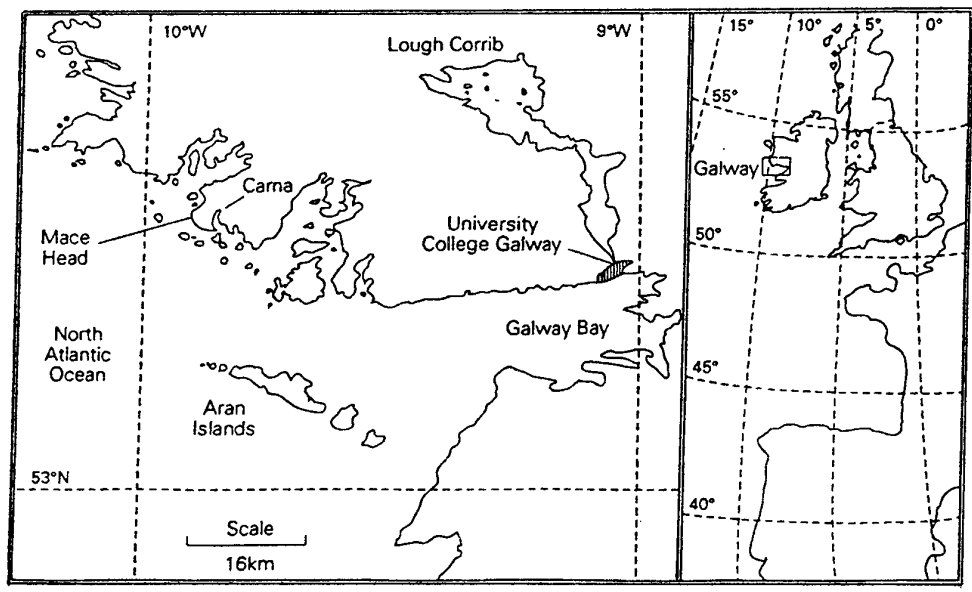
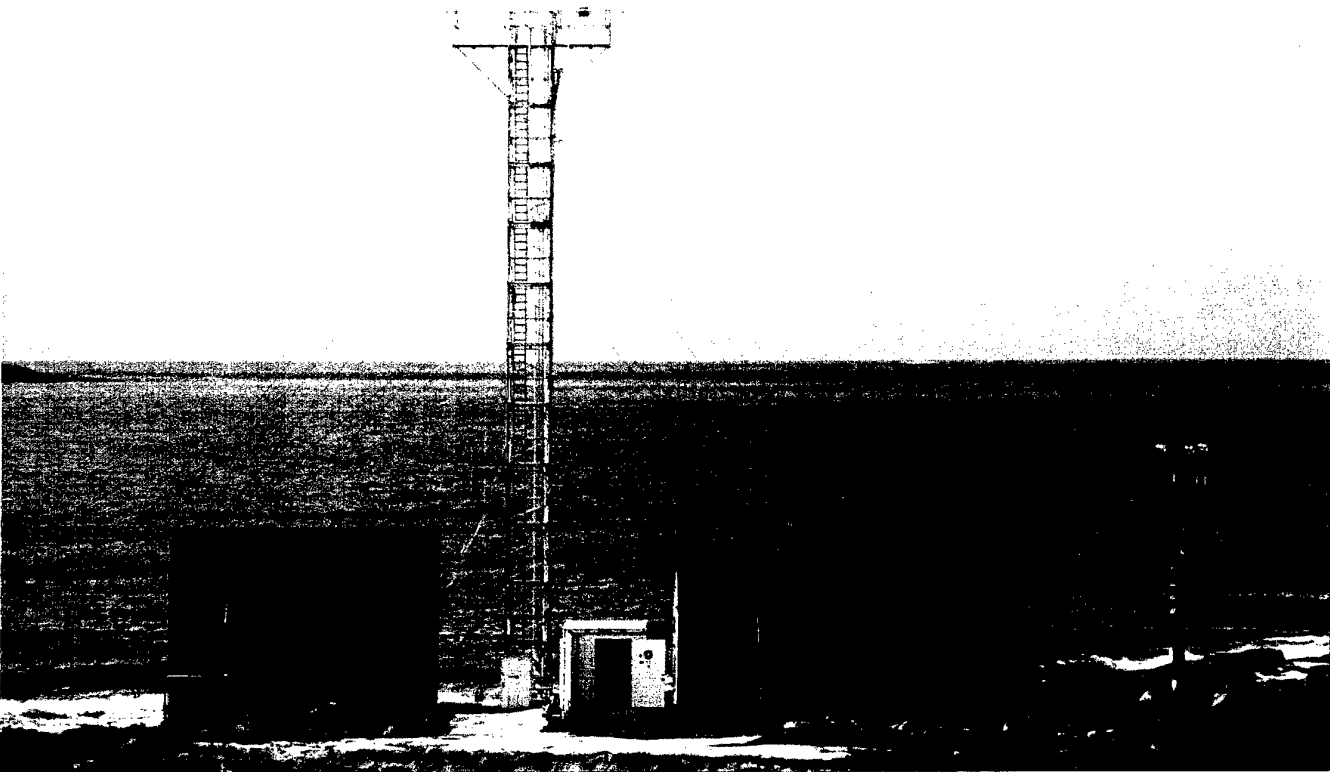
Measurements were made at Mace Head, Co. Galway (53°19'N, 9°54'W) of mass distributions together with aerosol volatility and supplementary meteorological parameters. This work (Jennings et al., 1996) extends earlier measurements at the site by Jennings et al. (1991) who provided particle size distribution spectra (in the absence of numerical data) for maritime, modified maritime and continental air masses. While the volatility of aerosol at Mace Head was first reported by Jennings and O'Dowd (1990), more emphasis was then placed on the extension of the technique as the inference of sodium chloride particles rather than the reporting of inferred number concentration of the aerosol.

The purpose of the field measurements is to investigate more fully and quantitatively the effect of air mass (in particular maritime and continental) on aerosol particulate levels at the relatively high latitude (53°N) Mace Head coastal site, representative of the mid North-Eastern Atlantic region.

A photograph of the two laboratory shore buildings at Mace Head, close to the sea-shore, together with a 20 m tower is shown in Figure 1. In addition, an extended cottage some 300 m from the shoreline and at an elevation 20 m above MSL was also used.

Maritime air is classified as air which arrives at the Mace Head site in a direction between 180° to 300° from the Atlantic Ocean. In addition to wind direction and air trajectory information, use is made of aerosol condensation nucleus (CN) number concentration to classify the air flow. CN number concentrations less than about 700 cm⁻³ together with airflow in the clean air sector are taken as indicators of maritime or marine air.

The site experienced three main maritime air flows: transatlantic maritime (M) (originating from a west-south west direction), tropical maritime (tM) (originating from a more southerly direction) and polar maritime (pM) (originating from a more



northerly direction). The transatlantic maritime (M) flow was the dominant maritime air flow encountered at the site. Air flows which originated over the ocean but which passed overland before reaching the site are classified as modified maritime (mM) air flows. Air coming from the UK or continental Europe is considered to be modified because of its traverse over the Irish sea and is classified as modified continental (mC) air. Selection of a particular air flow was based on a study of the combination of air flow trajectory data, local wind direction and CN number concentration at the site. Meteorological parameters such as wind speed, wind direction, temperature, relative humidity, precipitation presence, precipitation intensity and barometric pressure were recorded as part of the Atmosphere/Ocean Chemistry Experiment (AEROCE). Relative humidity is relatively high at the Mace head site, with a typical range of variability between about 75% - 85%. 925 mbar backward 4 day trajectories to Mace Head were calculated at the Meteorological Institute in Oslo and made available by Jozef Pacyna of the Norwegian Institute for Air Research. The 925 mbar value (corresponding to an altitude of 750m above MSL) was taken as a representative air flow trajectory within the atmospheric boundary layer.

Aerosol mass concentrations reported in this work were collected using a Berner low-pressure impactor with a flow rate of $30 \text{ dm}^3 \text{ min}^{-1}$. The impactor classified aerosol species in the diameter range between $0.06 \mu\text{m}$ - $16 \mu\text{m}$, where the sizes refer to the aerodynamic equivalent diameter (AED). Cutoff characteristics and geometric mean AED values are given in Table 2. The impactor was mounted on a tower positioned at approximately 300 m from the shore line at a height of 30 m above sea level. A computerised system facilitated sector control allowing differentiation between maritime and anthropogenically influenced air masses by means of wind direction data and CN particle levels. The impactor was set to sample in-sector air only, i.e. air masses within the designated clean sector of 180° - 300° .

Aerosol samples were collected on pre-weighed polycarbonate impactor foils and stored in pre-cleaned petri dishes prior to use. The foils were weighed before and after sampling using a $1 \mu\text{g}$ Sartorius microbalance under fairly constant conditions of relative humidity between 65% and 75%. Accumulation mode mass concentrations (\leq

geometric mean diameter of 0.71 μm) were calculated from mass concentrations calculated from impactor stages 1 to 4 inclusive.

Impactor stage	1	2	3	4	5	6	7	8
Aerodynamic diameter (μm)	0.06-0.125	0.125-0.25	0.25-0.5	0.5-1	1-2	2-4	4-8	8-16
Geometric mean AED	0.087	0.177	0.353	0.707	1.41	2.83	5.66	11.31

Table 2. Characteristics of 8-stage Berner Impactor in terms of the Aerodynamic Diameter and Geometric Mean Aerodynamic Diameter (AED)

A summary of the analysis of the microphysical data for the winter and summer seasons is given in Table 3. The data, Julian day, period of measurement and air mass type are given for each set of data. The values presented in Tables 3(a) and 3(b) represent mean values taken over the particular number of hourly cycles for the stated measurement dates. Total aerosol particle number concentration (cm^{-3}) and total volume ($\mu\text{m}^3 \text{ cm}^{-3}$) are presented primarily for maritime air. As a contrast to the cleaner air masses, microphysical data is also given for representative continental (modified) air masses encountered at Mace Head for both the winter and summer periods.

Through the use of volatility analysis, particle number and volume concentration for the chemical species of sulphuric acid, ammonium sulphate, sodium chloride and elemental carbon were inferred and are also presented in Table 3. Representative temperature fractionation curves for marine and continental air masses are shown in Figure 2 for a range of particle sizes. Both air masses show evidence for the presence of sulphuric acid (which volatilises at about 100°C). Ammonium sulphate shows a fairly sharp drop-off in number concentration, particularly for the smaller sized particles, at about 180°C . The marine aerosol number concentration shows a definite fall off, particularly for the largest size range, close to 600°C , which is indicative of the presence of sodium chloride particles. The sharper fall off in particle concentration at around $720^\circ - 730^\circ\text{C}$ for continental air for the two smallest size intervals is due to the presence of elemental carbon (Jennings et al., 1994).

3(a) Winter period.

Date	Julian Day	Time	Air mass Category	Total Number	Total Volume	Sulphuric Acid Number	Sulphuric Acid Volume	Amm. Sulphate Number	Amm. Sulphate Volume	Sea Salt Number	Sea Salt Volume	Elemental Carbon Number	Elemental Carbon Volume	Black Carbon (ng m ⁻³)	CN	CCN [†]
1/12/93	335	335.7 - 336.0	M	117	2.3	--	--	--	--	--	--	--	--	35.1	403	75
3/12/93	337	337.3 - 337.8	tM	108	1.3	--	--	--	--	--	--	--	--	18.3	358	95
4/12/93	338	338.6 - 339.0	pM	57.6	1.0	7.2	0.27	9	0.13	10.4	0.1	1.8	0.03	16.5	287	78
5/12/93	339	339.4 - 339.8	M	134	3.0	29.6	0.28	16.3	0.24	7.4	0.66	0.8	0.02	31.6	419	164
6/12/93	340	340.0 - 340.5	M	128	2.7	26.6	0.36	14.4	0.2	6.7	0.08	0.5	0.01	9.2	411	152
7/12/93	341	341.0 - 341.5	M	73.5	0.9	15.3	0.12	8.3	0.06	3.8	0.03	0.3	0.003	5.3	167	105
19/2/94	50	50.2 - 50.5	mM	174	3.0	36.2	0.42	19.6	0.21	9	0.1	0.7	0.01	185	2348	527
20/2/94	51	51.0 - 51.4	mM	198	3.1	41.2	0.43	22.3	0.22	10.2	0.1	0.8	0.01	172	2231	496
21/2/94	52	52.4 - 52.8	mC	1039	7.3	164	1.63	705	2.74	29.1	0.07	36.2	0.13	543	6942	1025
22/2/94	53	53.4 - 53.8	mC	1267	8.1	199	1.81	860	3.05	35.5	0.08	44.1	0.14	488	7539	1591
3/3/93	62	62.0 - 62.6	M	64.4	0.8	10.3	0.06	24.4	0.24	10.3	0.12	1	0.02	8.4	398	121
4/3/94	63	63.3 - 63.7	tM	72.2	0.9	12.3	0.10	23.6	0.19	6.3	0.08	2	0.01	8.6	931	249
6/3/94	65	65.6 - 66.0	M	126	2.3	21.5	0.27	41.2	0.51	11	0.22	3.5	0.03	11.4	427	184
7/3/94	66	66.3 - 66.8	M	164	1.4	31.2	0.16	51.4	0.25	15.4	0.4	5.1	0.06	6.5	341	82
8/3/94	67	67.0 - 67.4	M	63.6	1.0	12.7	0.07	17.4	0.25	7.2	0.24	0.4	0.02	5.4	497	204
9/3/94	68	68.0 - 68.4	M	80.3	0.9	22.7	0.16	16.6	0.14	4	0.45	1.1	0.02	5	305	69

Table 3(a). Accumulation mode aerosol microphysical and physico-chemical parameters measured at Mace Head over (a) the winter period and (b) the summer period in 1993 - 1994. Aerosol number concentration and aerosol chemical species number concentration is given in units of cm⁻³. Aerosol volume concentration and aerosol chemical species volume concentration is given in units of μm³cm⁻³. CN and CCN number concentrations are given in units of cm⁻³.

[†] CCN number concentrations for the winter period were measured at a supersaturation of 0.3%.

3(b) Summer period.

Date	Julian Day	Time	Air mass Category	Total Number	Total Volume	Sulphuric Acid Number	Sulphuric Acid Volume	Amm. Sulphate Number	Amm. Sulphate Volume	Sea Salt Number	Sea Salt Volume	Elemental Carbon Number	Elemental Carbon Volume	Black Carbon (ng m ⁻³)	CN	CCN*
23/5/94	143	143.6 - 144.0	mC	679	4.4	95.7	1.05	418	1.85	18.8	0.04	22.2	0.09	--	4418	542
24/5/94	144	144.4 - 145.0	mC	724	6.5	138	2.15	364	2.79	31.8	0.28	24.8	0.14	--	8457	1507
31/5/94	151	151.2 - 151.4	M	87	0.9	19.2	0.09	58.1	0.28	0.5	0.06	1.3	0.01	--	396	154
4/6/94	155	155.5 - 155.8	mM	173	2.4	35.9	0.03	56.1	0.16	6.4	0.07	0.8	0.05	--	1294	482
5/6/94	156	156.6 - 157.0	M	234	3.0	36.9	0.23	75.1	0.02	18.4	1.37	3.1	0.02	--	407	104
6/6/94	157	157.0 - 157.5	M	198	1.9	33.8	0.03	57.1	0.14	11.8	0.26	2	0.03	--	313	101
21/7/94	202	202.0 - 202.5	M	135	1.6	40.3	0.06	28.8	0.23	6.9	0.8	2.1	0.03	--	497	94
22/7/94	203	203.0 - 203.5	M	358	2.8	74.2	0.37	102	0.4	31.8	0.71	2.4	0.03	34.8	483	156
24/7/94	205	205.3 - 205.6	M	96	1.0	19.8	0.08	23.3	0.23	10.4	0.31	0.7	0.02	42.5	792	167
25/7/94	206	206.6 - 207.0	M	232	2.7	42.6	0.29	83.2	0.45	21.1	0.16	8	0.05	78.3	887	257
26/7/94	207	207.6 - 208.0	M	146	2.1	22.9	0.63	24.5	0.25	27.3	0.27	4.8	0.08	52.6	711	239
28/7/94	209	209.0 - 209.2	M	78	1.0	12.3	0.29	13.2	0.11	14.7	0.12	2.6	0.04	35.2	287	34
29/7/94	210	210.7 - 210.9	mM	47	0.6	7.3	0.19	7.9	0.07	8.7	0.08	1.5	0.03	86.4	528	137
5/8/94	217	217.0 - 217.4	M	39	0.7	6.1	0.21	6.6	0.08	7.3	0.09	1.3	0.03	21	348	25
12/8/94	224	224.0 - 224.4	M	89	1.0	25	0.16	40.3	0.07	5.1	0.25	0.3	0.09	23.6	319	20
14/8/94	226	226.1 - 226.4	M	177	1.9	30.4	0.17	124	0.16	--	--	--	--	50.6	503	34

Table 3(b). Accumulation mode aerosol microphysical and physico-chemical parameters measured at Mace Head over (a) the winter period and (b) the summer period in 1993 - 1994. Aerosol number concentration and aerosol chemical species number concentration is given in units of cm⁻³. Aerosol volume concentration and aerosol chemical species volume concentration is given in units of μm³ cm⁻³. CN and CCN number concentrations are given in units of cm⁻³.

* CCN number concentrations for the summer period were measured at a supersaturation of 0.5%.

Temperature Fractionation Curves

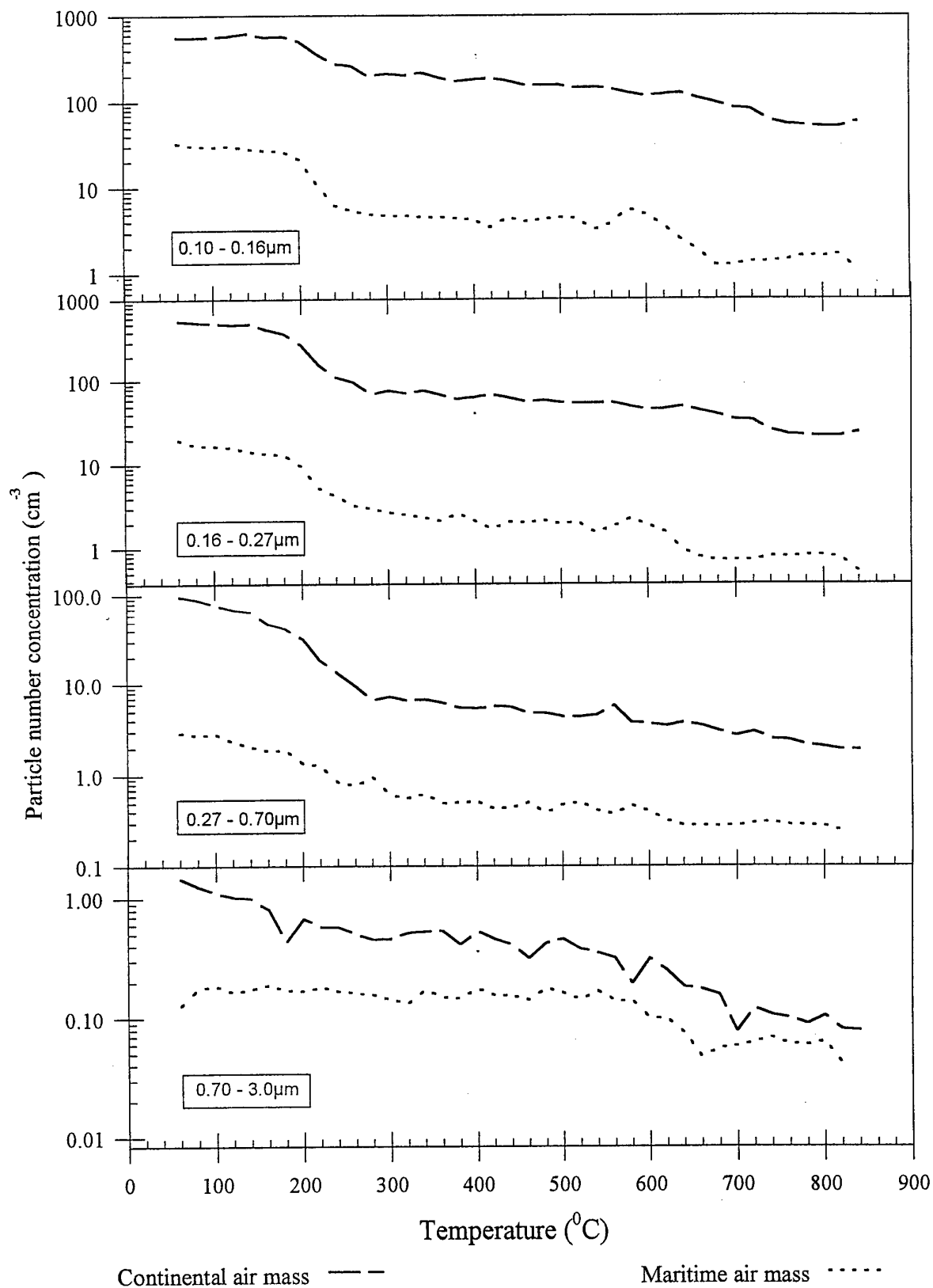


Figure 2. Temperature fractionation curves for field data recorded at Mace Head for maritime and continental air, for four particle diameter size ranges.

In addition, averaged values of condensation nuclei (CN), cloud condensation nuclei (CCN) and black carbon (BC) are also presented in Table 3 for the same measurement periods as those used for the aerosol sizing/volatility events. The levels of both CN and CCN are clearly a sensitive indicator for characterising the two main air masses. CCN concentration values of about 100 cm^{-3} typifies marine air conditions, with values a factor of 10 higher for continental air. It can be seen that the CCN/CN ratio is greater (0.23 - 0.34) for marine air compared to about 0.17 for polluted air.

4.2 Aerosol Mass Concentration Measurements at Mace Head

Measured aerosol mass concentrations for in-sector marine air samples taken at Mace Head in 1993 and 1994 (Jennings et al, 1996) are shown in Table 4. The mass data are averaged over the winter and summer seasons. Total mass concentrations measured during the summer are somewhat higher ($29.4 \pm 5.6 \mu\text{g m}^{-3}$) than those taken during the winter period ($23.5 \pm 5.4 \mu\text{g m}^{-3}$). The accumulation mode mass of $5.5 \mu\text{g m}^{-3}$ (for both seasons) contributes 24% and 18% of the total mass for the winter and summer seasons respectively.

(b) Season	Mean total mass concentration ($\mu\text{g m}^{-3}$)	Mean accumulation mode mass concentration ⁺ ($\mu\text{g m}^{-3}$)
Winter	23.5 ± 5.4	5.5 ± 1.2
Summer	29.4 ± 5.6	5.4 ± 1.0

⁺ Aerosol mass collected in the first 4 stages of the Berner impactor, diameter $\leq 0.71 \mu\text{m}$

Table 4. Total Aerosol Mass Concentration and Accumulation Mode Mass Concentration for Marine Air for Winter and Summer Periods

The distribution of aerosol mass in accordance with aerosol aerodynamic diameter is shown in Figure 3 for marine air. Mean values of $dM/d\log D$ together with standard errors for winter and summer seasons are presented. Both data sets show a bimodal form with a more pronounced bimodal behaviour for the summer measurements. Fitted log-normal distribution parameters for the mass data together with calculated mass based on those parameters are given in Table 5.

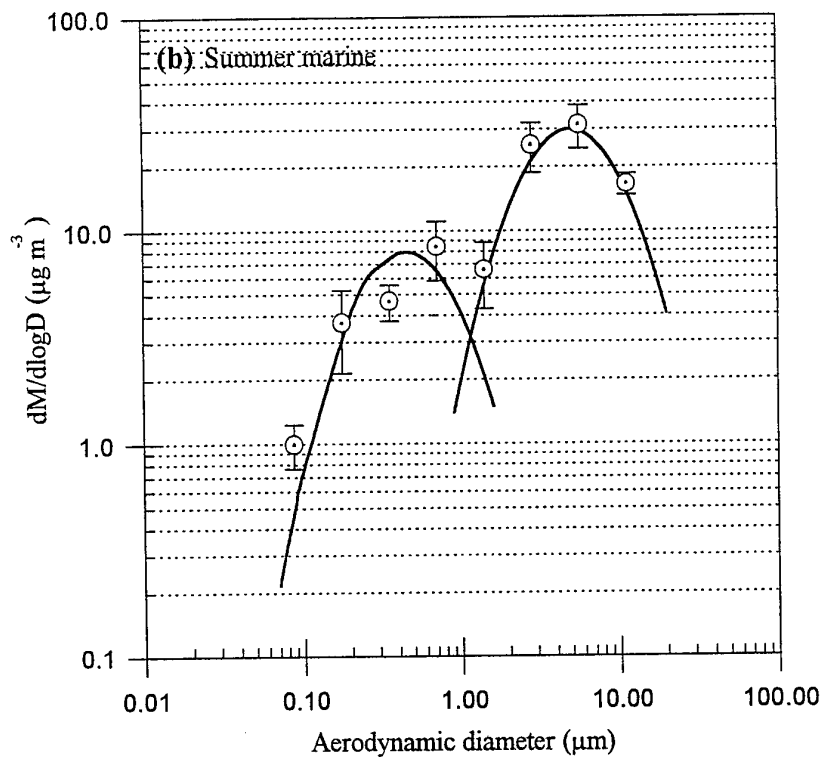
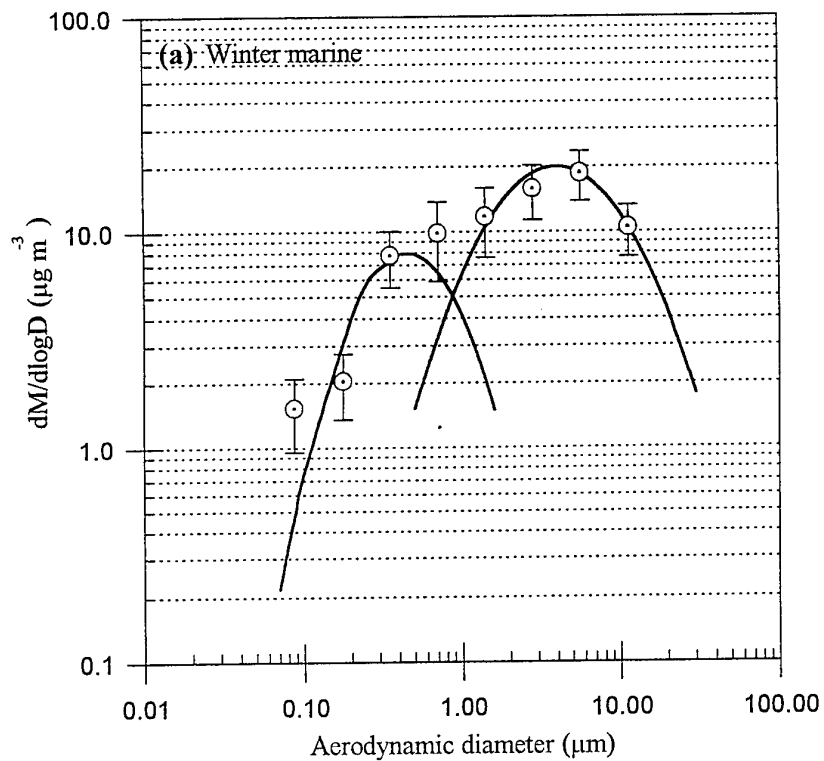


Figure 3. Mass distribution of marine air for 1993 - 1994 at Mace Head, (a) Winter season, (b) Summer season. Log-normal fitted curves (with parameters given in Table 5) are superimposed in the data.

Season	Accumulation Mode				Coarse Mode			
	D_m μm	σ_g	P_m $\mu g m^{-3}$	$Mass^+$ $\mu g m^{-3}$	D_m μm	σ_g	P_m $\mu g m^{-3}$	$Mass^+$ $\mu g m^{-3}$
Winter	0.4-0.5	2.0	8.0	6.0	4.0	2.5	20	20.0
Summer	0.4-0.5	2.0	8.0	6.0	5.0	2.0	30	22.6

Table 5. Log-Normal Mass Distribution Parameters Fitted to the Impactor Mass Measurements for Marine Air in 1993-1994 at Mace Head

D_m is the geometric mean mass and σ_g is the geometric standard deviation.

P_m is the peak value of the $dM/d\log D$ versus D curve (Figure 3).

$Mass^+$ is that calculated using the tabulated log-normal parameters.

4.3 Black Carbon Aerosol Volatility

A volatility technique whereby aerosol particles are heated to the relatively high temperature of 860°C is used to infer the presence of elemental carbon in polluted air masses at Mace Head. Aerosol volatility to date, including the work of Pinnick et al. (1987), Clarke et al. (1987), Jennings and O'Dowd (1990), has permitted inference of at least three main atmospheric aerosol constituents: sulphuric acid, ammonium sulphate and sodium chloride. This section briefly describes the extension of the volatility technique to that of elemental carbon in carbonaceous aerosols and a fuller description is given by Jennings et al. (1994).

Laboratory volatility measurements of carbon and carbonate aerosols were first carried out to determine the form of characteristic fractionation curves for a range of carbonaceous aerosols. The laboratory data is then used as a reference data set with which to compare ambient aerosol fractionation curves in order to infer the presence of elemental carbon.

Aerosol volatility measurements made at Mace Head during the period 27-28 May 1992 revealed a significant reduction in particle number concentration for particle radii up to about 0.2 μm , between temperatures 110°C and 860°C, which is attributed to the presence of elemental carbon. The volume and percentage volume for the inferred volume (over the diameter range from about 0.1-0.84 μm) is 0.28 $\mu m^3 cm^{-3}$ and 4.2% respectively. This estimated percentage (of between 2-4%) found in this work is in

broad agreement with the findings of some other workers such as Stevens et al. (1984) and Heintzenberg and Covert (1984).

5. MEASUREMENTS OF BIOLOGICAL AEROSOL AT MACE HEAD

5.1 Measurements of Biological (Pollen and Spore) Aerosol

The Burkard volumetric trap was used to obtain pollen and spore grain concentration per m^3 of air at Mace Head. Pollen and spore counts are combined for the months of March through June 1993 and are shown in Figure 4. Total pollen and spore count m^{-3} for the periods of July to December 1993 and January to June 1994 are shown in Figures 5 and 6.

The effect of air mass on bioaerosol concentration was examined. Marine air, from the clean air sector, yielded zero concentration of bioaerosol as shown in the 7th interim report. In contrast, both modified continental and maritime air masses result in typical levels of bioaerosol measured at the site.

The measurement and analysis of bioaerosol using passive Tauber traps is summarised in the 7th interim report. Taxa recorded at Mace Head are shown in Table 6.

Within 5m of the platform:		At a distance of more than 5m:	
Taxon	Abundance	Taxon	Abundance
Ericaceae	a	Solidago	o
Calluna	a	Lythrum	o
Potentilla Type	f	Jasione montana	o
Plantago Lanceolata	f	Chrysanthemum	o
Molinia caerulea	o	Plantago coronopus	f
Poa pratensis	f	Plantago Lanceolata	f
Lotus	f	Plantago maritima	f
Carex	f	Festuca	a
Succisa	o	Gramineae	a
Plantago maritima	o		
Centaurea nigra	o		
Ulex	o		
Rumex acetosa	r		
Salix	o		

Table 6. Taxa observed at Mace Head

Figure 4

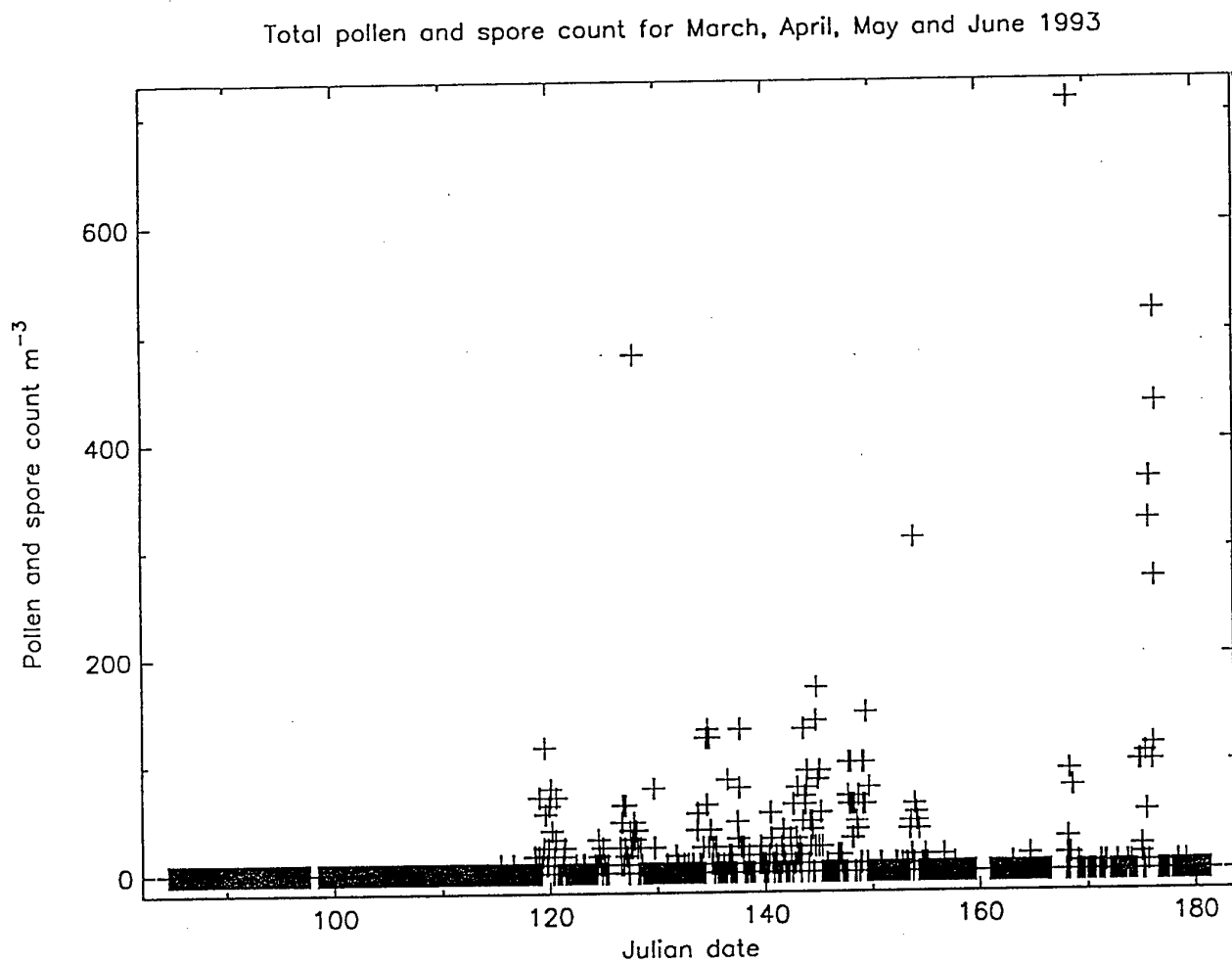


Figure 5

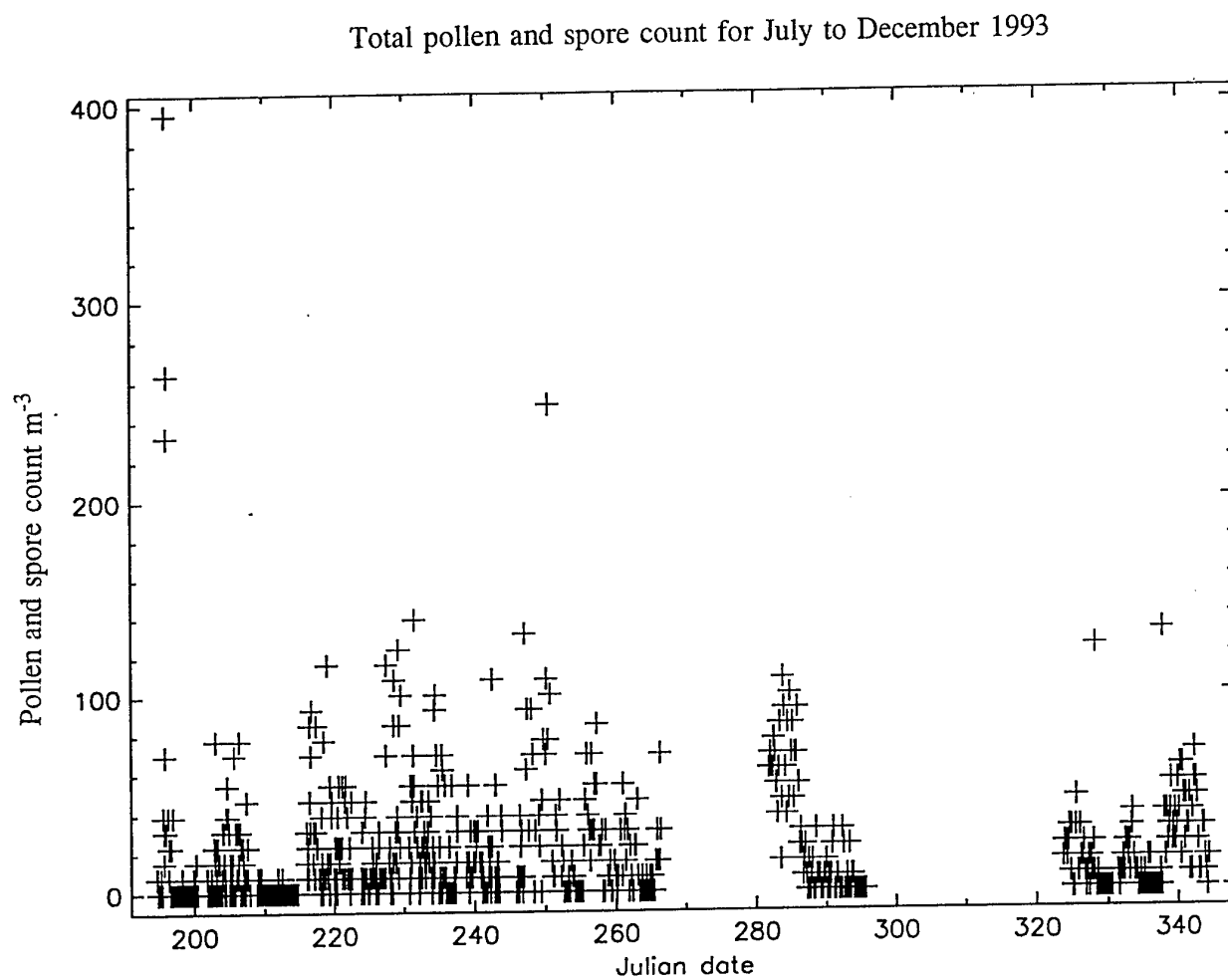
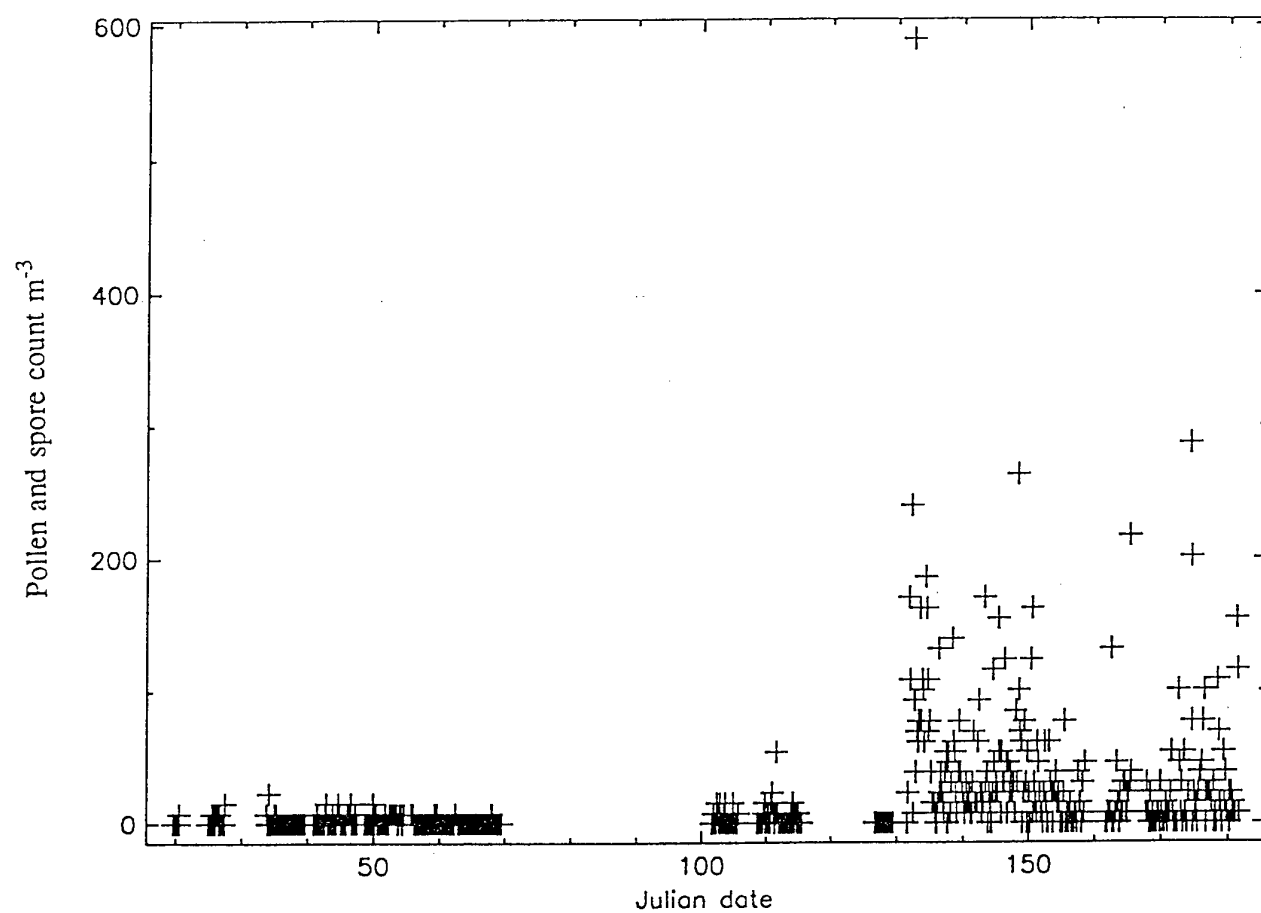


Figure 6

Total pollen and spore count for January to June 1994



Their abundance is rated as follows: dominant=d, abundant=a, frequent=f, occasional=o, rare=r. Percentage values of taxa in each sample as well as concentration influx (number of pollen grains cm^{-2} per day) are presented in the 7th interim report.

5.2 Comparison of the Sampling Methods

The Tauber trap is a useful instrument for obtaining particle information. The trap itself is a convenient device because changeover of traps is quick and simple. The results obtained for the Tauber trap show such a small concentration of biological aerosols that it would perhaps be an improvement to change the trap less often. The purpose of this work was to observe the deposition on a shorter time scale, which is why the method used was chosen. The frequent trap changes were employed also because of the enormous quantity of precipitation at the sites. The techniques involved in sample preparation cause the biological particles to become quite visible using the microscope, but some particles are at risk of destruction by acetolysis. The sample is occasionally marred by the intrusion of insects but this is impossible to avoid completely using any particle collector. The locations chosen were remote and were chosen partly on the basis of their remoteness to observe situations influenced by the Atlantic ocean and also to minimise interference by passers by.

The Burkard volumetric trap is a useful technique for observing airborne particles by impaction. The samples obtained can be examined to a precision of two hourly intervals. The aerobiological content of the atmosphere is often small at the Mace Head Field Research Station, where the Burkard trap was positioned. However, this method provides information on very low concentrations. It is theoretically possible using this technique, to identify all pollen and spores visible with a microscope, because at no stage in the preparation are the grains at risk of destruction. It would be extremely time consuming to identify every known taxa and most research in this area involves the examination of relatively few taxa for a given time interval. The impacting particles are undamaged by any preparation steps, which means that every particle is visible, the only hindrance to this capability occurs when the view of one particle is obstructed by another particle. The identification of particles is more difficult than in the case of the Tauber trap samples because the particles are fixed and cannot be

turned or rotated. The difference in visibility without acetolysis is obvious when viewing slides. The Burkard trap placement at Mace Head had the advantage of regular observation. Any problems concerning the instrument could be recorded and corrected within a short space of time. The Burkard trap would not be suitable for monitoring in a completely remote site, as access needs to be gained weekly to facilitate changing the Burkard drum. Insects did cause an interference with the sample, as in the case of the Tauber traps, but with the Burkard trap this did not cause as much data to be lost. Storms or strong winds would resuspend particles from the ground having a detrimental effect on the accurate analysis of samples obtained, but this cannot be avoided by any method of aerobiological sampling.

5.3 Bioaerosol Sampling Using Glass Cyclone Systems

5.3.1 Review of glass cyclone samplers

Effective biological analysis of airborne particles requires samplers operating at a high flow rate and with the capability of concentrating the air particulate into a fairly small liquid volume. The glass cyclone sampler fulfills these two requirements. The flow pattern in the return flow tangential inlet cyclone, used here, is described by Ogawa (1984). A comprehensive evaluation of biological aerosol samplers is reviewed by Henningson and Ahlberg (1994). Errington and Powell (1969) were the first to design a cyclone sampler where the microorganisms are collected in a liquid. The cyclones had collection fluid continuously sprayed into the sampler inlet and operated at flow rates of 75 and 350 L per minute.

A large volume (of order 1000 L per minute or less) is described by Decker et al. (1969). The principle of operation, which applies to the (glass) cyclone in general, is based on the production of a fine liquid mist in a rapidly moving airstream with collection of the aerosol in the liquid which is collected at the cyclone outlet. Collection efficiencies averaged 70% for *S. marcescens* 1 μm size aerosol organisms.

Since the cyclone was at that time being developed by Aerojet General Corporation, under Government contract, it became known as the Aerojet General cyclone. This cyclone sampler was further described by Buchanan et al. (1972). The airborne

particles are removed from the airstream mainly through impingement into the liquid film formed along the cyclone walls, and then washed off into a collecting reservoir. The collection efficiency of the Aerosol General cyclone was measured by May et al. (1976) and more recent collection efficiency work by Upton et al. (1993) and Griffiths et al. (1993) extend the range of aerosol size up to 20 μm . A summary of collection efficiency values obtained for the glass cyclone sampler is shown in Table 7.

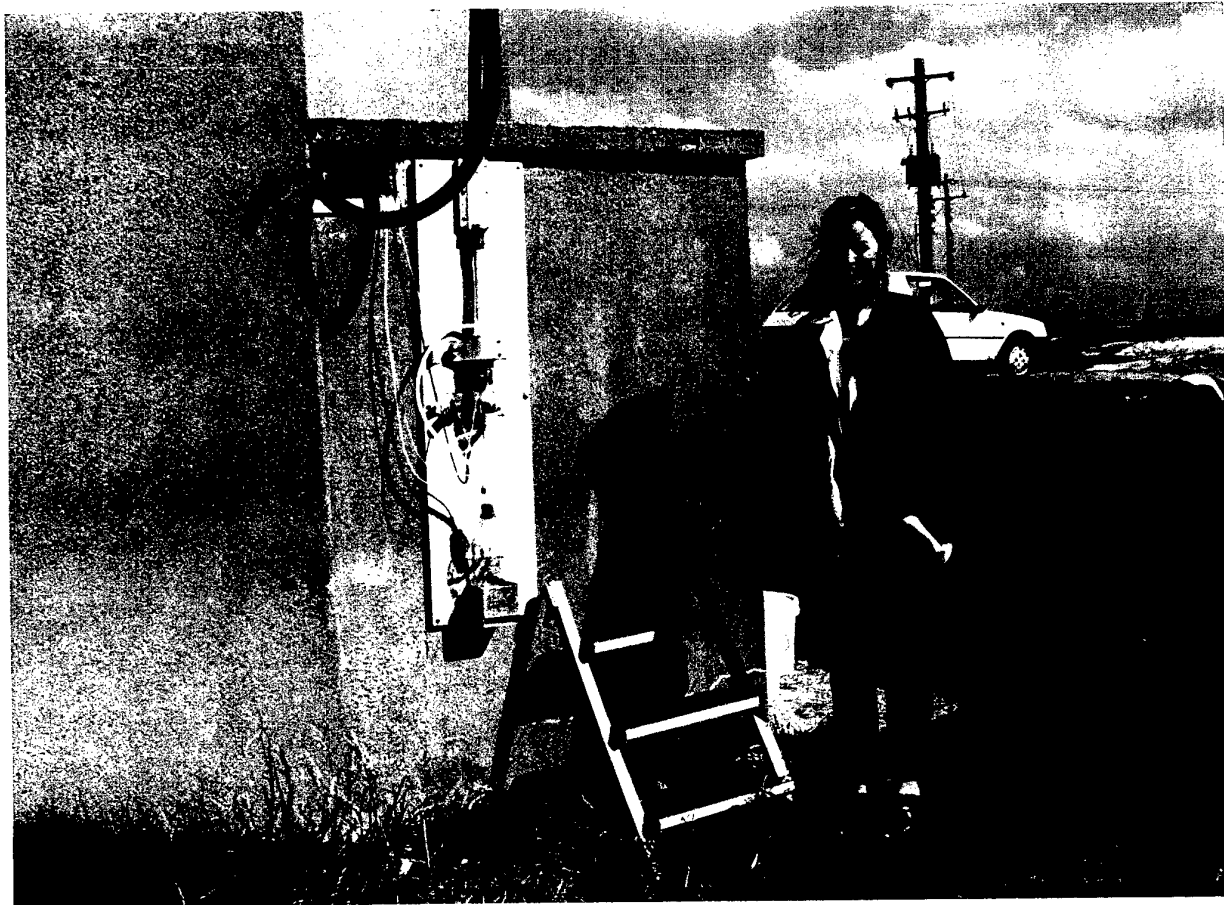
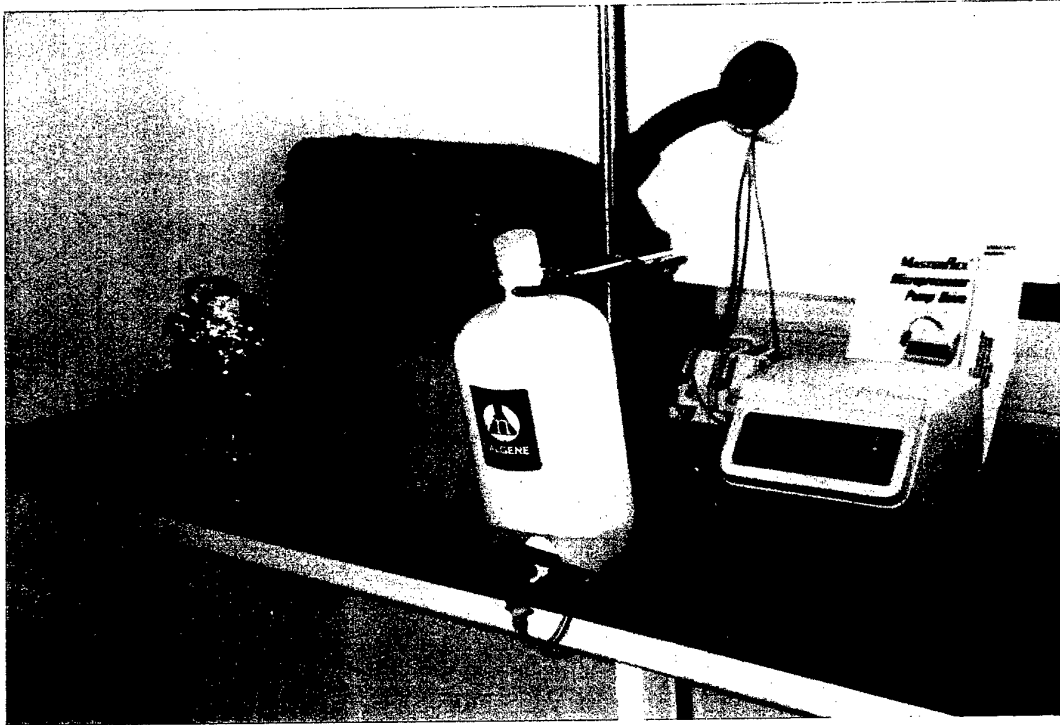
5.3.2 Glass Cyclone Sampler System Used in this Work

The glass cyclone sampler is available from the Hampshire Glassware Company (Griffiths & DeCosemo, 1994). Two such cyclones have been procured for this work. In addition a 1300 watt (1.75 hp) Electrolux variable speed vacuum cleaner, with the capability of aspirating air @ 500 L per minute has also been purchased. A rotameter is placed in the suction line between the cyclone and the vacuum cleaner - this is considered to be a reliable and direct method of monitoring the flow rate. A digitally controlled peristaltic pump (manufactured by Ismatec, UK Ltd.) has the capability of delivering flow rates between 0.06 and 3.6 mL per minute when used with size 13 silicone tubing. The microprocessor controlled unit accurately permits the desired flow at a rate of 1 mL per minute. Two pump heads are used - one to deliver the buffer solution to the cyclone and - the other to deliver the collected sampler solution to the collecting bottle. The peristaltic pump system and a photograph of Caroline Kenny with the glass cyclone system is shown in Figure 7.

It should be mentioned that with the prevailing relatively high humidity values (typically in excess of around 70%) at the background field station at Mace Head on the west coast of Ireland, that evaporative losses of buffer solution are fairly minimal. The protocol for set-up, cleaning and sterilizing the glass cyclone sampler, used at ERDEC, as developed and written by Sarah Cork has been provided to University College Galway.

Table 7 Collection efficiency values for the glass cyclone

Bioaerosol Species	Mean diameter (1 μm)	Wind Speed ms^{-1}	Collection Efficiency(%)	Work references
<i>S. marcescens</i>	1	0(laboratory)	70	Decker et al (1969)
<i>Bacillus subtilis</i> var <i>niger</i>	0.9	0 (laboratory)	71	Buchanan et al (1972)
	1.8	0 (laboratory)	90	Buchanan et al (1972)
	3.6	0 (laboratory)	96	Buchanan et al (1972)
<i>S. cerevisiae</i>	20	1 - 6	66	May et al (1976)
	30	1 - 6	74	
	5 - 20	0 - 4	80 - 100	Upton et al (1993)



5.3.3 Bioaerosol Fluorescence Measurement Method

Total fluorescence of the bioaerosol samples is determined using a Perkin Elmer LS 50B fluorometer available at University College Galway. The protocol used for fluorescence measurements at ERDEC by Dr. Steven Christesen and Kate Ka Ong has been adopted for use in this work. Samples collected for total fluorescence measurements are analysed within 24 hours. The sampling media used is phosphate buffered saline pH 7.4 (Sigma Diagnostics, 120 mmol/L NaCL), mixed with 1 L distilled water. Further description is given in the 8th Interim report.

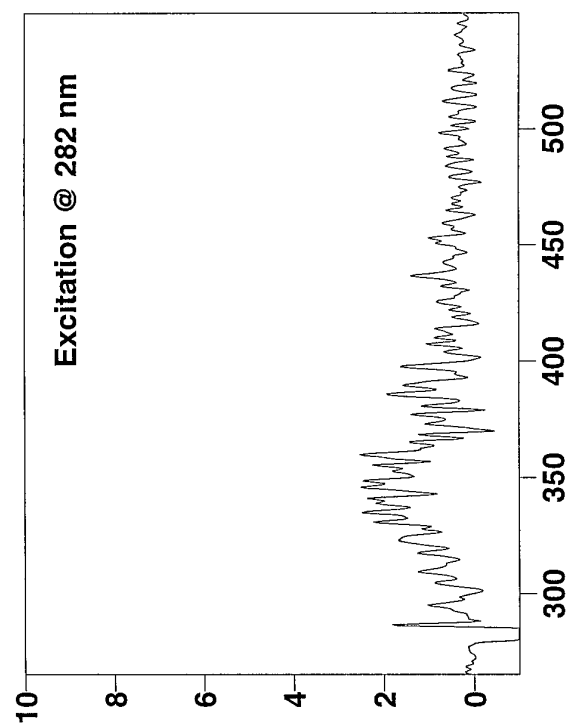
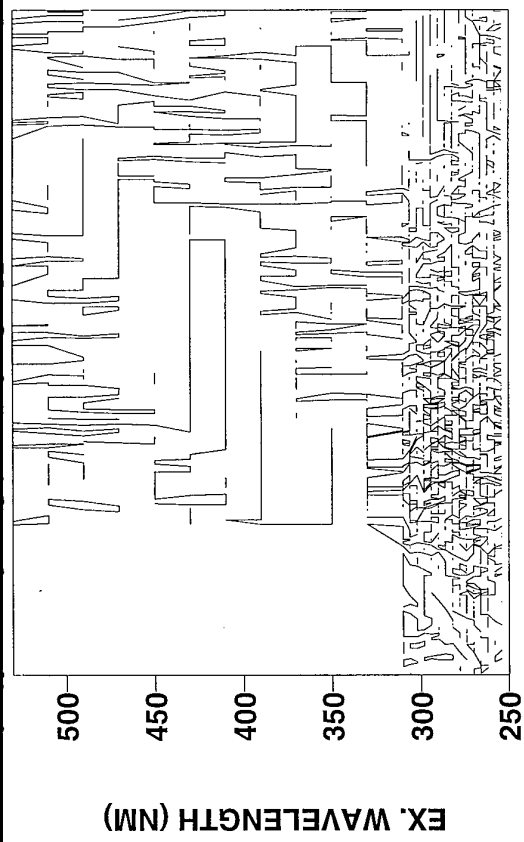
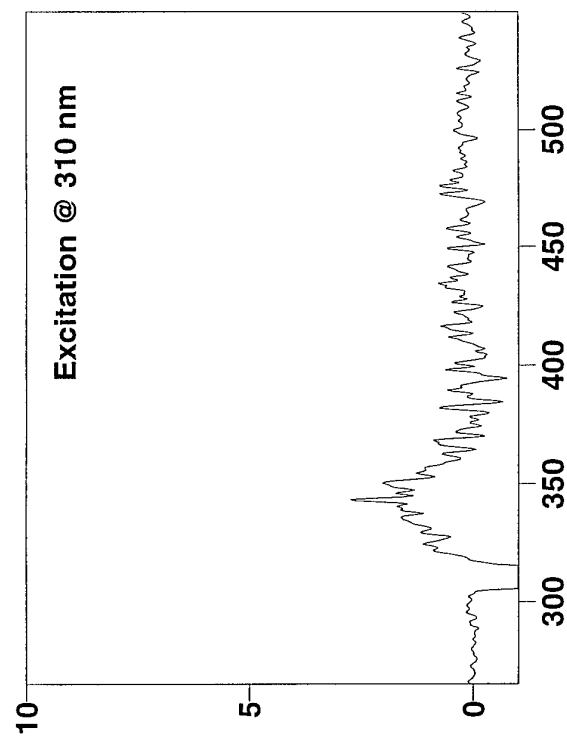
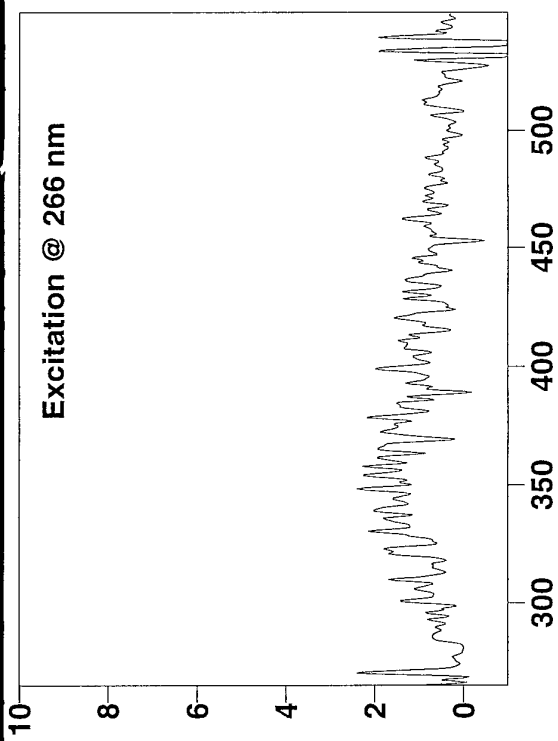
The Perkin Elmer spectrofluorometer is used over the excitation wavelength range of 250 - 310 nm, in 4 nm steps and emission wavelength from 265 - 550 nm, in 1 nm steps for scheme I and excitation range 310 - 530 nm, in 20 nm steps, emission wavelength from 330 - 800 nm, in 1 nm steps for scheme II. Representative spectra obtained from bioaerosol samples with the glass cyclone system at Mace Head are shown in Figure 8 (Dr. Steven Christesen, private communication, 1996).

5.4 Background Aerosol Characterisation

Preliminary background field analyses of the natural aerosol particle size distribution measured (using a TSI aerodynamic particle sizer) over the same period as the biological sampling, have been carried out. Representative measurements of size distribution for marine air (background) and polluted air masses are shown in Figures 9 and 10. A comparison of the size spectra shows that the particulate number concentration dominates for the marine aerosol in the 1 - 10 μm diameter range as might be expected.

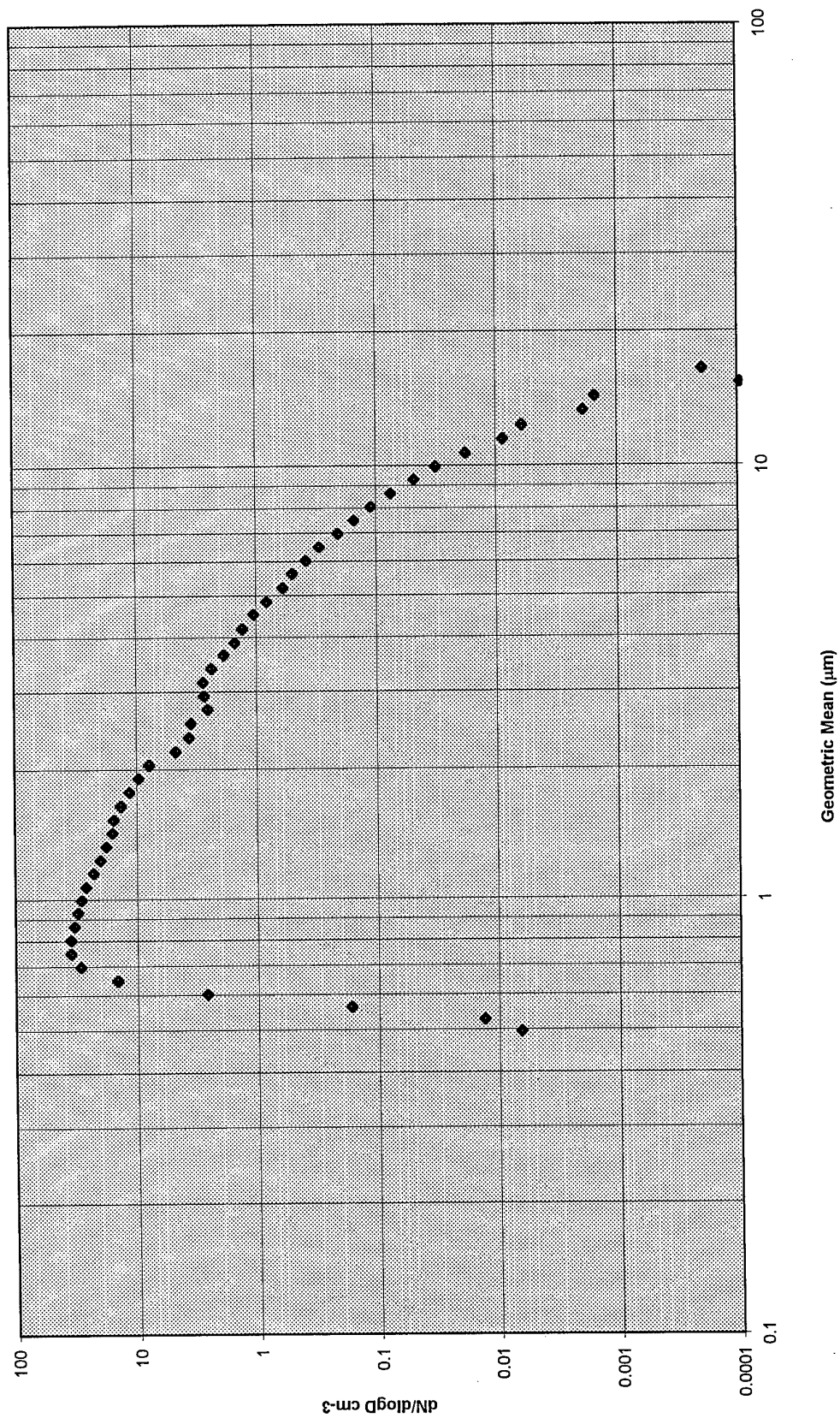
6. ACKNOWLEDGEMENTS

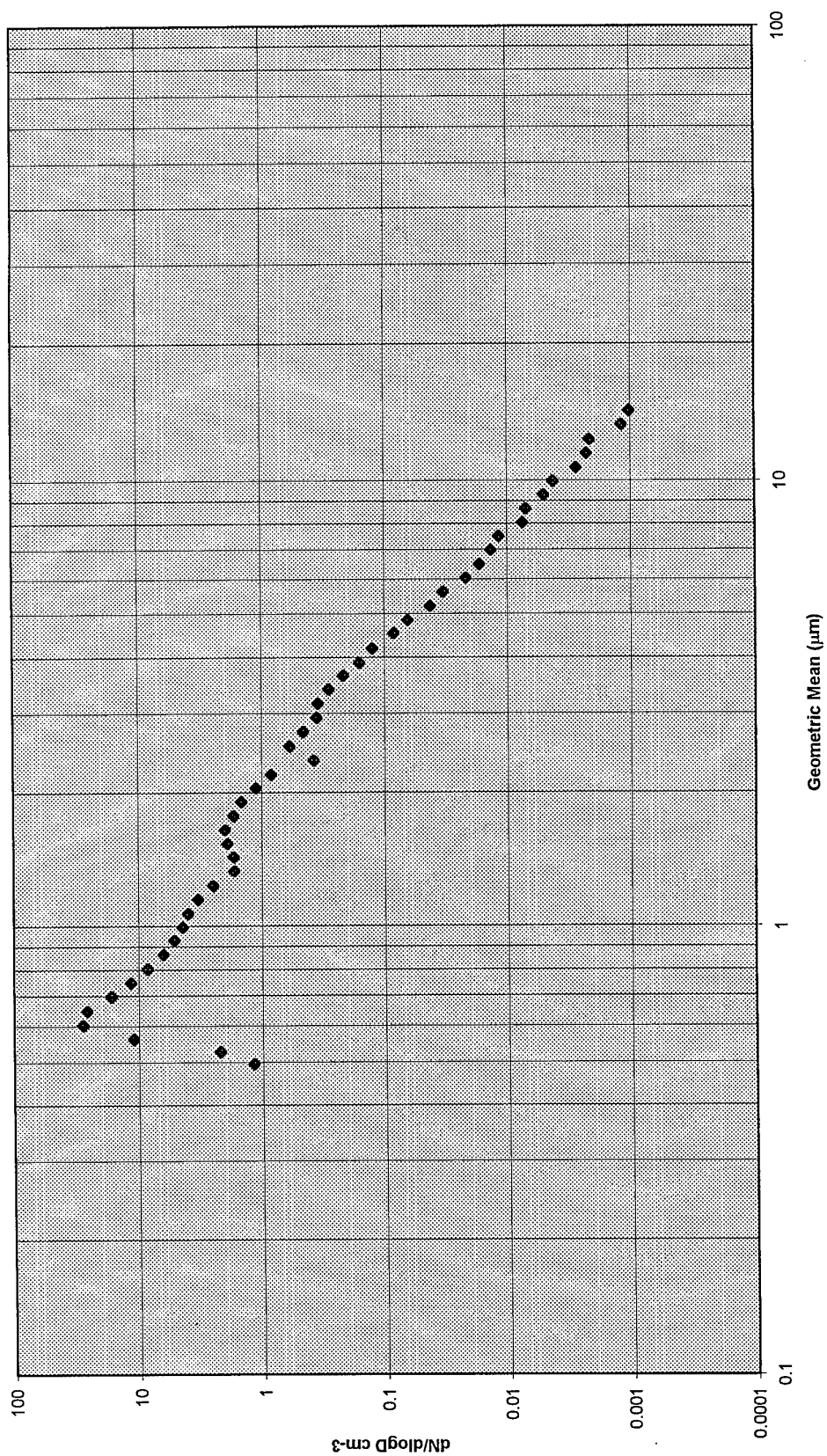
We wish to acknowledge the much appreciated support of Dr. Steven Gotoff, Standoff Directorate Division for this work. The cooperation of the ERDEC team - Dr. Hugh Carlon, Dorothy Paterno, Sarah Cork, Dr. Steven Christesen and Kate Ka Ong, together with their Branch Chief Charles Wick and Director Patrick M. Nolan are also greatly appreciated. This work has been supported by the European Research Office, US Army Research, Development & Standardisation Group, London, England through contract number: DAJA-45-92-C-0024.



EMISSION WAVELENGTH (NM)

71096S with Water Subtracted





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